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LAMINATING METHOD OF BIAXIALLY DRAWN POLYESTER FILMS
[Nijiku ensho poriesuteru fuirumu no hariawase hoho]

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Claim

A laminating method of biaxially drawn polyester films characterized by the fact that two biaxially drawn polyester films are laminated by thermocompression to each other such that the angle between their principal orienting axes is 40° or smaller.

Detailed explanation of the invention

The present invention pertains to a laminating method of biaxially drawn polyester films.

More specifically, the present invention provides a laminating method characterized by the fact that biaxially drawn polyester films are laminated to each other under prescribed conditions so that a thermocompressed laminate appropriate for use as the base of a magnetic storage card with excellent flatness can be obtained.

Among plastic films, polyester film has the highest strength, and it has excellent elongation and impact strength. It is stable in a wide temperature range, and it has high chemical resistance, that is, it can withstand various types of organic solvents, oils and fats, and acids. Also, it has excellent electric properties and transparency. Due to said excellent physical and chemical properties, high heat resistance, transparency, and good electrical properties, the polyester film has been in wide use as photographic film base, magnetic tape base, and electrical insulating material. In addition, as one of the recent new applications, it can be used as the magnetic storage card base in the form of laminate.

For example, it is used as the base of the individual identification card, that is, magnetic debit card for automatic teller machines, credit card, and other magnetic storage cards.

For the laminated sheet made of polyester film for said application, usually, a laminating agent is coated on one side of the polyester film, and the laminated sheet is formed by means of thermocompression for laminating. The following schemes may be adopted in applying a laminating

layer on one side of the polyester film: in one method, as the laminating agent, polyethylene, ethylene/vinyl acetate copolymer, ethylene/acrylate copolymer, ionomer resin or the like is kept in melt state, while coating is performed by means of doctor blade method, roll coater method, or the like. In another method, an extruder is used to extrude [the melt] from a flat die to form a film, and thermocompression is performed while in the melt state for laminating. In yet another method, a solution prepared by dissolving in an organic solvent is coated and dried to form a laminating agent layer on one side of the polyester film. Then, with the laminating agent layer as the internal layer, thermocompression is performed for laminating to form the laminated sheet.

When the laminating agent is coated, as needed, an appropriate anchoring treatment may be performed beforehand.

As an example, in the following, an explanation will be given in more detail regarding the case when it is used for the holding sheet of a magnetic debit card. In this case, a magnetic layer having the necessary items stored in it is formed on one side of the polyester film, and a laminating agent layer is formed on the other side. Then, a display piece with necessary items printed on it is set on the laminating agent layer. Then, another polyester film having no magnetic layer and only a laminating agent layer is laminated on it, followed by thermocompression to form the magnetic debit card for automatic teller machine.

In this case, there are some problems. The obtained sheet prepared by laminating may develop twist as a deformation (twist), or one layer may bend inwards (curl). In such case, the flatness of the laminated sheet degrades, so that troubles may take place when it is inserted into an automatic teller machine. Especially, it is difficult to correct the twist, so that the trouble becomes serious. Improvement has to be made.

The present inventors have performed extensive research on the cause of the twist of the laminated sheet, and have found that an important factor is the molecular orienting angle of the polyester films used as the base [material]. As a result, the present invention was reached.

The present invention provides a laminating method of biaxially drawn polyester films characterized by the fact that two biaxially drawn polyester films are laminated by thermocompression to each other such that the angle between their principal orienting axes is 40° or smaller.

In order to guarantee good flatness for the laminated sheet obtained by thermocompression laminating processing, it is necessary to ensure balance of the state of thermal dimension variation of the films in laminating in the heating process and cooling process, that is, in the heating/cooling process for the thermocompression laminating processing. If this balance is lost, bending deformation takes place on either side, leading to twist or curl.

The present inventors have performed extensive research on this phenomenon. As a result, it has been found that the relative position of the orienting angles of the principal molecular orientation in the facing portions of the laminated base films is an important factor that causes the aforementioned phenomenon. Figure 1 is a diagram illustrating the results of the experiment indicating the relationship.

Figure 1 is a diagram illustrating the results of measurement of the curl degree as the deformation generated in the laminated ribbon-shaped films, which have a width of 10 mm and have different angles formed between the principal orientation and the longitudinal direction, for determining the model of the relationship between the principal orienting directions in the facing portions of the laminated base films and the deformation of the laminated sheets.

Here, the curl degree is represented as the curvature (1/radius (m^{-1})) of the arc when the laminated films bend inwards with respect to one side.

In Figure 1, the abscissa represents sin θ, the angle formed between the principal molecular orienting axes, a parameter of the relative position of the principal molecular orienting directions of the two laminated films.

As can be seen from Figure 1, in the thermocompression laminating processing operation, due to the bending deformation proportional to angle sin θ between the principal molecular orientations of the laminated films in the thermocompression laminating processing, when thermocompression laminating is performed to form a sheet shape, bending deformation takes place to one side due to the angle formed between the molecular orientation of the facing portions. In a synthesized form, the deformation leads to variation in the flatness of the laminated sheet, and, according to the degree and distribution of such variation, twist or curl takes place. The molecular orientation property of the biaxially drawn polyester film mainly depends on the drawing conditions in the longitudinal and lateral directions and the thermocompression condition. For the biaxially drawn film manufactured by the successive biaxial drawing method using the tenter scheme as a conventional manufacturing method, due to the bowing phenomenon, while lateral drawing takes place correctly in the lateral direction of the film when the film is manufactured, in the end portions of the film, drawing in the lateral direction deviates from the correct direction. Consequently, although the principal orienting direction of molecules is parallel or perpendicular to the film manufacturing direction in the film manufacturing operation, it is inclined in the film end portions.

In this way, the molecular orientation in the film plane of the biaxially drawn polyester film is usually non-uniform in the various directions in the plane. As a result, in order to obtain the laminated sheet with excellent flatness, it is necessary to realize balance in the dimensional variation quantity by laminating the films such that their principal orienting axes of their portions facing each other are parallel to each other.

In the practice, for the magnetic debit cards, usually, the films are slit in the longitudinal direction, and the ribbons are laminated to each other, followed by cutout. Consequently, as described in Japanese Kokoku Patent Application No. Sho 48[1973]-38775, only the central portion of the film where the principal molecular orienting axis is relatively parallel to the longitudinal or lateral direction of the film is used so as to ensure less significant bowing phenomenon.

Another scale of the orientation property in the film plane of the biaxially drawn polyester film is the birefringence. For a film with birefringence of 0, there exists no principal orienting angle. On the other hand, for the biaxially drawn polyester film prepared by successive drawing method that is conventionally adopted, usually, birefringence is not zero. According to the method of the present invention, irrespective of the value of the conventional birefringence, it is possible to obtain the laminated sheet with excellent flatness.

When a laminated film is used in forming the magnetic debit card for automatic teller machine, with respect to the flatness that is especially important, there is the twist as the longitudinal deformation. As a scale of the flatness, when the twist is measured, it is preferred that the twist quantity be 5% or smaller, or more preferably 3% or smaller, for the practical application.

Here, the twist quantity is represented by the value in units of mm measured for the distance of floating of a corner among the four corners of the sheet from the flat plate caused by twist when the thermocompression laminated sheet in rectangular shape measuring 55x70 mm is set on the flat plate.

The deformation with twist over 5 mm depends on the thickness of the laminated sheet. Usually, in the in-plane direction of the laminated base films, it is caused by the thermal dimensional variation quantity of 0.02% or more at the corresponding positions, respectively.

Consequently, it is necessary to restrict the angle formed between the principal molecular orienting axes of the base films within a prescribed range. That is, the angle between the principal molecular

orienting axes of the laminated films, while depending on the molecular orientation property, should be 40° or smaller, or preferably 28° or smaller, so as to ensure that no twist takes place to cause problems in the practical application. In this way, the purpose of the present invention is realized.

By restricting the angle between the principal orienting axes of the laminated base films within the aforementioned range, it is possible to obtain a laminated sheet with excellent flatness. The reason is as follows: for the biaxially drawn polyester films, variation in dimensions due to thermal causes includes two types, namely, thermal shrinkage, an irreversible variation, and thermal expansion/shrinking, a reversible variation. In the thermocompression laminating processing operation using a hot melt type laminating agent, at the temperature higher than the softening point of the laminating agent at which the laminating agent can easily make deformation, even when variation in dimensions of the laminated films takes place, the buffering function of the laminating agent layer acts so that deformation takes place independently, while the overall flatness of the laminated sheet is not degraded. Also, the reversible thermal shrinkage of the biaxially drawn polyester films, which have excellent dimensional stability among the plastic films, can be ignored at the temperature lower than the softening point of said hot melt type laminating agent. Consequently, it is believed that the important factor for maintaining the flatness in the thermocompression laminating processing is not the thermal shrinkage, an irreversible change. Instead, it is the change in dimensions caused by the variation in temperature in the temperature range lower than the softening point of the laminating agent, that is, the reversible expansion shrinkage due to thermal expansion, and the main factor dominating the thermal expansion rate in various directions in the plane of the film for the homogeneous polyester film that is manufactured continuously is the orientation property of the molecules.

The polyester film in the present invention is polyethylene terephthalate copolymer, with most of the repeating units of polyethylene terephthalate and containing a small quantity of other dibasic groups or diol.

The laminated sheet manufactured according to the present invention is used in manufacturing magnetic debit cards, credit cards, etc. Of course, it can be used in applications that do not need the magnetic layer of coating.

In the following, an explanation will be given in more detail regarding application examples of the present invention.

Application Example 1

Biaxially drawn films were formed with different angles of the principal orienting axis by changing the longitudinal and lateral drawing rate and drawing condition of a substantially undrawn polyester sheet prepared by extruding a melt of polyester resin from dies and quenching it. Each of the obtained biaxially drawn films was used in the following test. The film is coated with a 35-μm-thick thermocompression laminating layer made of ethylene-acrylate copolymer (DQOA-2609 manufactured by Union Carbide Corp.) by means of extruding coating. Each film is then cut to cards (55x70 mm) with the longitudinal direction of each card parallel to the major edge. Two obtained cards are then laminated with the adhesive layer on the inner side by means of thermocompression at 130°C and under a pressure of 1 kg, followed by cooling. The characteristics of the obtained cards are listed in Table 1.

TABLE 1. Angle (θ) between the principal orienting axes of the two laminated films and the card characteristics

① 試料No.	② 主配向の角度	θ	③ ツイスト	④ 使用判定
1	0°	0°	0	○
2	[illegible]0	[illegible]	0	○
3	15	30	26	○
4	20	40	50	○～△
5	25	50	4[illegible]	×
6	[illegible]	[illegible]	[illegible]	×
7	45	90	[illegible]	×

⑤ 使用判定
○ 使用出来る
△ 場合により使用出来る
× 使用不可

⑥ 主配向角度：フイルムの長手方向に対する主配向の角度

Key: 1 Sample No.

2 Principal orienting angle

3 Twist

4 Grade of use

5 Grade of use:

O: Can be used

Δ: Can be used in some cases

X: Cannot be used

6 Principal orienting angle: Angle of the principal orientation with respect to the longitudinal direction of the film

<u>Application Example 2</u>

Biaxially drawn polyester films were manufactured by performing longitudinal and lateral drawing using the tenter method for a substantially undrawn polyester sheet prepared by extruding polyester resin melt from dies, followed by quenching. For each obtained biaxially drawn polyester film, the central

portion has a principal orienting angle of 2°, while the end portions have a principal orienting angle of 36°.

The central portion and end portion of the film are used respectively in laminating performed according to the method described in Application Example 1. It is found that for the laminated card prepared from the central portion of the film manufactured, the crossing angle between the principal orienting axes of the two films is 4°, and there is no twist. The result is good. On the other hand, for the sample prepared from the end portion, the crossing angle between the principal orienting axes of the two films is 72°, and the twist is 7.5 mm. Consequently, it cannot be used in this case.

In this way, for the polyester films manufactured under the same condition, if the crossing angle between the principal orienting axes is large, the obtained laminated card develops twist too large to find any practical application.

Application Example 3

Using sample No. 2 obtained in Application Example 1, cards are cut out at various different angles between the principal orienting direction and the major edge of the card. Then, for each type of card, laminating is performed according to the method described in Application Example 1. In this case, the relationship between the angle θ of the principal orienting direction of the films on the outer/inner sides and the card characteristics are listed in Table 3. When θ is over 50°, no sheet with excellent flatness can be obtained.

TABLE 3. Relationship between θ and card characteristics

1 試料No	θ	2 ツイスト	使用判定 3
1	0	0	◎
2	20	1.3	○
3	40	4.1	○
4	50	5.0	○~△
5	70	8.1	×
6	90	9.2	×

Key: 1 Sample No.

2 Twist

3 Grade of use

As can be seen from the aforementioned application examples, in order to obtain laminated sheet with excellent flatness, it is necessary to ensure that the relative position of the principal orienting angle of the laminated films is within the crossing angle defined in this invention.

Usually, for the magnetic debit cards, etc., as the laminated sheet prepared by thermocompression laminating polyester films is cut out parallel to the longitudinal direction or lateral direction of the base films, in order to obtain laminated sheet with excellent flatness all the time, when laminating is performed in ribbon shape, it is necessary to ensure that the base films have appropriate orienting angle with respect to the longitudinal or lateral direction of the mainly molecular orientated film such that the crossing angle between the principal orienting axes becomes within the range of the present invention.

Brief description of the figures

In Figure 1, the ordinate shows the curl degree as the variation quantity of the ribbon shaped laminated film when films with width of 10 mm are thermocompression laminated, and the abscissa

shows sin θ, the angle between the principal orienting axes of the films, as the parameter of the relative position of the principal orienting angle of the two laminated films.

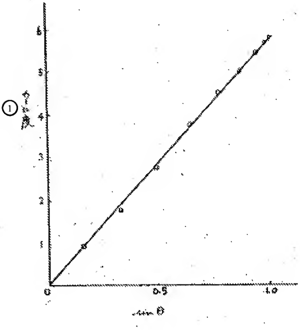

Figure 1

Key: 1 Curl degree